

A COMPILATION OF INVENTORIES OF EMISSIONS  
TO THE ATMOSPHERE

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**Abstract.** Detailed and accurate emissions inventories are essential for reliable computer dispersion model simulation of the behavior of chemically and radiatively important atmospheric species. Currently, model simulations of the atmosphere are limited by the paucity of quality emissions data for input. As a first step in providing internationally recognized emissions inventories, we list here the inventories that are extant, together with their spatial and temporal characteristics and a few interpretive comments. The only global emissions inventory we regard as good is that for chlorofluorocarbons. Those for CO<sub>2</sub>, CH<sub>4</sub>, NO<sub>x</sub>, SO<sub>2</sub>, reduced sulfur, and radon we regard as fair. In selected

regions, the spatial resolution of emissions is well determined for CO<sub>2</sub>, CO, NO<sub>x</sub>, and SO<sub>2</sub>. The temporal resolution of existing inventories is almost uniformly poor. Much remains to be done to generate emissions inventories adequate to fully support computer models of regional and global chemistry and climate.

OVERVIEW

Assessments of the status and possible future of air quality and atmospheric chemistry are based in part on estimates of the fluxes of species emitted into the atmosphere: the so-called "emissions inventories." Inventories are required for a variety of uses, and, in consequence, have been prepared at a variety of spatial resolutions, species detail, and accuracy. Many inventories are not publicly documented or readily available, characteristics which have encouraged the development of new inventories rather than the use of those already prepared.

Emissions inventories play an interactive role with atmospheric measurements and with model studies. In one mode of operation, emission inventories are used as input conditions for models, and the model results are validated by comparison with ambient observations. In another, measurements are used as input conditions to the models, and the model results then provide an estimate of emissions. In a third, the emissions inventories and model results suggest locations or times when measurement activities would be particularly important or instructive. The three independent approaches to atmospheric chemistry and its driving forces are thus intrinsically linked, and provide the potential for mutual validation.

With many computer models being used to predict the future of ozone, global warming, and other important policy issues, model verification becomes a matter of central importance. In such efforts, the use of common, reliable

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emissions inventories eliminates one possible source of disagreement among models. However, few global inventories have won general acceptance by the modeling community.

These considerations emphasize the need for emissions inventories, present and projected, that are regarded by atmospheric scientists as authoritative and reliable and that are readily available to prospective users. Accomplishing this goal is the ambitious task of the Global Emissions Inventories Activity (GEIA), a component of the International Global Atmospheric Chemistry Project (IGAC). For all species of interest, the GEIA goal is to develop and/or certify emissions inventories on a  $1^\circ$  by  $1^\circ$  grid (throughout this paper, the first dimension indicates latitude, the second longitude). As a prelude to that task, we have assembled a compilation of existing inventories for atmospheric gases and particles, and for associated supporting data.

Inventories are constructed in two distinct ways. One mode is the process inventory, in which all emissions from a specific process, such as biomass burning or volcanic eruption, are assessed. The other mode is the species inventory, in which the assessment is done for emissions of a specific compound or group of compounds from all sources. Both types of inventory are important, and most inventory activities are inevitably a combination of both types. For tractability, however, the present publication deals only with species-type inventories.

We realize that a compilation of inventories will inevitably be incomplete and will become quickly dated. Nonetheless, its existence will enable modelers to review and utilize existing inventories as the efforts to prepare and certify the GEIA components proceed. We invite those familiar with information not cited here to communicate the details to us.

## PROCEDURES FOR INVENTORY DEVELOPMENT

All emissions inventories ultimately seek to compute emission rates from a formula of the type

$$E_{i,\Delta t,A_{x,y}} = \sum_t \sum_{A_{x,y}} \sum_s N_{s,A_{x,y}} U_{s,A_{x,y},t} \Phi_{s,i,t} \quad (1)$$

where

- $E_{i,\Delta t,A_{x,y}}$  is the emission rate of species  $i$  over time interval  $\Delta t$  over an area specified by the coordinate  $x$  and  $y$ ,
- $N_{s,A_{x,y}}$  is the number of sources of type  $s$  in area  $A_{x,y}$ ,
- $U_{s,A_{x,y},t}$  is the time-dependent usage given to source  $s$  in area  $A_{x,y}$ ,
- $\Phi_{s,i,t}$  is the flux of species  $i$  from source  $s$  at time  $t$ . Flux terms are sometimes called "emission factors."

The basic step in inventory construction is, of course, to decide which species are of interest, and what geographical area is of concern. These two decisions determine much of the subsequent complexities and procedures. If methane emissions around the world are of interest, for example, every  $A_{x,y}$  on the globe must be treated and some way must be found to count (or estimate) the number of each type of methane source in such different regions as Northern Europe,

the mid-Pacific Ocean, and Antarctica. The result is generally that the land areas used are large and the time intervals are large as well, probably annual. Conversely, if carbon monoxide emissions in a large city are of interest, the variety of sources will not be large. Time intervals are likely to be daily or hourly, and the metropolitan region will be the only area of interest.

Once the species and area have been determined, the sources of species  $i$  can be identified and counted. For nitrogen dioxide in an urban area, for example, these include anthropogenic sources such as motor vehicles, power plants, and industrial operations, and natural sources such as lightning. Motor vehicles need to be subdivided into automobiles of different types, weights, and fuel efficiencies, as well as trucks, buses, and so forth, because each produces a unique amount of  $\text{NO}_2$  per unit of fuel.

When sources are identified and counted, it is then necessary to determine how much and how often they are used. For example, it might be necessary to determine automotive travel frequency for each hour of the day or each day of the week. For natural sources, we might wish to determine such factors as vegetative growth rates in each season of the year.

Those who determine all of the  $N_{s,A_{x,y}}$  and  $U_{s,A_{x,y},t}$  values are performing an accounting function of great complexity. Also, they are almost inevitably forced to make assumptions or estimates when information is absent. For regional anthropogenic  $\text{NO}_x$ , for example, it is common to have the number of sources incompletely known, operating parameter information incomplete, and industrial process emissions unspecified. Notwithstanding these difficulties, unless the accounting is carefully and accurately done, the resulting inventory will suffer significantly in accuracy.

The evaluation of the flux terms,  $\Phi_{s,i,t}$ , is an exercise not in accounting, but in science, engineering, or both. The central question is the rate of emission of species  $i$  given a particular source and set of conditions (type of fuel, ambient temperature, etc.). The answer to this question may involve in situ sampling of industrial processes, field measurements of natural processes, and the like. The process is as complex as the accounting functions involved in inventory development but requires different professionals with different skills.

It is appropriate also to mention the interactions of emissions inventories with computer models. Most inventories are generated because they are to be used as input to such models, and the limitations and aims of the models often dictate the form of the supporting inventories. Conversely, models can sometimes serve as tools for the estimation of emissions. This occurs when a model reproduces atmospheric concentrations of a species of interest, computes its rates of loss (to surfaces, chemical transformations, the stratosphere, etc.), and finds a requirement for a total emission rate that may differ from previous assessments. In this way, theory and observation interact to produce a more accurate result than would occur by either acting alone.

## SUPPORTING DATA BASES

At  $1^\circ \times 1^\circ$  resolution, the surface of Earth is divided into  $\approx 15,000$  ice-free land grid boxes,  $\approx 7000$  land-ice grid boxes,

and  $\approx 43,000$  ocean grid boxes. It is unlikely that there will ever be direct measurements at each grid box for each source function of each trace gas. It is more likely that direct flux measurements will be made at a few "representative" sites and for a few "representative" processes, and that relationships will thus be established between the measured fluxes and a suite of controlling properties. The global distributions of these controlling properties will form the framework for extrapolating point measurements in space and time to yield global estimates of the emissions.

In this section and in Table 1 we present a sampling of the global data bases useful for the modeling of trace gas emissions. The sampling is by no means exhaustive but is chosen to illustrate the wealth of information already at hand. The information is divided into primary data bases, which describe the spatial and temporal variations of atmospheric, terrestrial, and oceanic properties that may control or modulate trace gas fluxes, and secondary data bases which describe anthropogenic activities that are important for trace gas emissions. Many of the data sets represent climatology averaged over an observing period. Others are snapshots in time or are time series useful for analyzing interannual and longer-term variations or for understanding atmospheric chemistry at a specific point in time.

#### *Primary Supporting Data Bases*

**Atmosphere.** The comparatively long history of observations and mathematical models of the atmosphere has generated a large number of gridded data bases of circulation statistics of the atmosphere. Three-dimensional distributions of wind, temperature, and humidity of the entire troposphere have been compiled from observations [e.g., Oort, 1983]. The Oort monthly climatology is based on atmospheric observations from 1958 to 1973 and is gridded at  $2.5^\circ \times 5^\circ$  resolution with 11 levels in the vertical.

Real-time observations are by their nature synoptic and sparse. In numerical weather prediction, these observations available every 12 hours are processed through a general circulation model (GCM) to produce filtered, gridded, and filled-in arrays referred to as "analyzed" data [Hoskins et al., 1989; Kalnay et al., 1990], which are treated as real-time observations of the atmosphere. As in any data processing, the accuracy of the data product is intimately tied to the analysis procedure, in this case the equations and parameterizations in the GCM [Trenberth and Olson, 1988]. Lau [1984] presented analyzed circulation statistics for 1979. Monthly and seasonal statistics for 1980–1987 on  $4^\circ \times 5^\circ$  global grid are summarized by Schubert et al. [1990a, b].

Near-surface climate is generally described in terms of monthly surface air temperature and precipitation [Shea, 1986]. The Shea climatology is based on weather station data from 1950 to 1979 and is gridded at  $2^\circ \times 2.5^\circ$  resolution. Comparable data sets are those of Legates and Willmott [1988] gridded at  $0.5^\circ \times 0.5^\circ$  resolution. Hahn et al. [1988] have constructed gridded cloud data from surface observations. Month-to-month variations in surface air temperature anomalies in the last  $\approx 100$  years have been compiled by Hansen and Lebedeff [1987, 1988], Jones et al. [1986a, b, c], and others. Globally averaged and

hemispherically averaged temperature anomalies ( $\Delta T$ ) are provided by Hansen and Lebedeff and by Jones et al. In addition, Hansen and Lebedeff estimated the  $\Delta T$  for 80 equal area boxes for the globe, and the anomalies data can be easily interpolated to any other grid, including  $1^\circ \times 1^\circ$ . Because of the small-scale variations of precipitation, there is as yet no comparable global gridded data on actual monthly precipitation values. Eischeid et al. [1991] present secular variations of "global" precipitation since 1880, the "global" averages being computed using all  $4^\circ \times 5^\circ$  grid boxes with precipitation observations.

The International Cloud Climatology Project (ISCCP) combines measurements from four geostationary and one or two polar orbiting satellites to produce global distributions of cloud cover, cloud optical thickness, cloud top temperature, cloud top pressure, surface temperature and surface reflectance [Schiffer and Rossow, 1983, 1985]. "Stage C1 data" of ISCCP represents three-hourly distributions of these parameters at 280 km resolution (equivalent to  $2.5^\circ \times 2.5^\circ$  at the equator) for the globe since July 1983 while "Stage C2 data" are monthly averages of the Stage C1 data (cf. Tables 1 and 2 of Rossow and Schiffer [1991]). Included in the ISCCP data sets are other atmospheric properties used in the ISCCP retrievals, for example, tropospheric temperature and water vapor profiles and column ozone. Derived from the "C1" data is the monthly mean incident solar insolation at the surface [Bishop and Rossow, 1991].

**Land.** For the land surface, the primary data bases include topography, vegetation, and soils, compiled mostly from classical survey maps. Gates and Nelson [1975] give topographic height and ocean bathymetry at  $1^\circ \times 1^\circ$  resolution for the globe. This data set is widely used to produce topographic variations in coarse-resolution general circulation models. A new high-resolution data set, ETOPO5 [National Geophysical Data Center (NGDC), 1988], combines bathymetry data from the U.S. Naval Oceanographic Office with land elevations obtained from remote sensing and national surveys. Although the grid spacing of the data base is  $5' \times 5'$ , the information is at  $5' \times 5'$  only for the oceans, the "lower-48" United States, Japan, Western Europe, New Zealand, and Australia, and is at  $10' \times 10'$  elsewhere [NGDC, 1988]. A data base for albedo is available [Matthews, 1983]; it is an important variable in climate models. Also of interest, particularly for energy studies, is a country-scale inventory of coal resources [Couch, 1988].

A companion to the topographic data base is that on river drainage basins [Russell and Miller, 1990]. Average direction of actual or potential water flow over the area of a  $2^\circ \times 2.5^\circ$  grid box is inferred from topographic maps. On the basis of direction, each grid box was associated with drainage into a particular river. The data have been applied to compute streamflows in general circulation models [Miller and Russell, 1990].

Global data bases of vegetation are obtained by digitizing regional and global vegetation maps with various scales. A commonly used data base is that of Matthews [1983, 1984] which discriminates, at  $1^\circ \times 1^\circ$  resolution,  $>170$  vegetation types classified by the hierarchical physiognomic classification scheme. A complementary but independent data set, compiled at the same time and resolution, includes information on dominant land use practice in each  $1^\circ \times 1^\circ$  grid

TABLE 1. Existing Global Primary Data Bases

Parameter	Spatial Resolution <sup>a</sup>	Temporal Resolution	Effective Year <sup>b</sup>	Reference
<i>Atmosphere</i>				
Vector wind	2.5°x5°x11L	monthly	(1958-1973)	Oort [1983]
Temperature	2.5°x5°x11L	monthly	(1958-1973)	Oort [1983]
Specific humidity	2.5°x5°x11L	monthly	(1958-1973)	Oort [1983]
Surface air temperature	2°x2.5°	monthly	(1958-1979)	Shea [1986]
ΔT surface	4°x5°	monthly	1880-1990	Hansen and Lebedeff [1987]
ΔT surface	hemisphere	monthly	1861-1990	Jones et al. [1986a, b, c]
Precipitation	2°x2.5°	monthly	(1958-1979)	Shea [1986]
Δ precipitation	"global"	seasonal	1880-1990	Eischeid et al. [1991]
Clouds	280 km x 280 km	3-hourly	1983-1990	Rossow and Schiffer [1991]
Clouds	5°x5°	monthly	1952-1981	Hahn et al. [1988]
Tropical temperature	280 km x 280 km x 5L	monthly	1983-1990	Rossow and Schiffer [1991]
Tropical water vapor	280 km x 280 km x 5L	monthly	1983-1990	Rossow and Schiffer [1991]
Total column O <sub>3</sub>	280 km x 280 km	monthly	1983-1990	Rossow and Schiffer [1991]
<i>Land</i>				
Topog/bathymetry	1°x1°	none	present	Gates and Nelson [1975]
Topog/bathymetry	5' - 10'	none	present	NGDC [1988]
Albedo	1°x1°	none	present	Matthews [1983]
Coal resources	Country	none	present	Couch [1988]
Vegetation	1°x1°	none	present	Matthews [1983]
Vegetation	0.5°x0.5°	none	present	Olson and Watts, 1982
Land cover	1°x1°	none	present	Wilson and Henderson-Sellers [1985]
Land use	1°x1°	none	present	Matthews [1983]
Soils	1°x1°	none	present	Zobler [1986]
Soils	1°x1°	none	present	Wilson and Henderson-Sellers [1985]
Wetlands	1°x1°	none	present	Matthews and Fung [1987]
Wetlands	2°x2.5°	none	present	Aselmann and Crutzen [1989]
Drainage basins	2°x2.5°	none	present	Russell and Miller [1990]
Vegetation index	1°x1°	monthly	1982-1990	Tucker et al. [1986]
<i>Ocean</i>				
Sea surface temperature	2°x2°	monthly	1985-1994	Reynolds [1988]
Sea surface temperature	[1024x512]	monthly	1988	Halpern et al. [1991]
Marine climate	2°x2°	monthly	1854-1979	Slutz et al. [1985]
Surface wind	2°x2°	monthly	1983	Woodruff et al. [1987]
Surface wind	[1080x540]	monthly	1988	Halpern et al. [1991]
Surface height	[1080x540]	monthly	1988	Halpern et al. [1991]
Temperature	1°x1°x33L	seasonal	(1900-1978)	Levitus, 1982
Salinity	1°x1°x33L	seasonal	(1990-1978)	Levitus, 1982
Oxygen	1°x1°x33L	seasonal	(1990-1978)	Levitus, 1982
Ocean color	[512 x 512]	seasonal	1978-1986	Feldman et al. [1989]
ΔpCO <sub>2</sub>	2°x2°	seasonal	(1972-1990)	Tans et al. [1990]

<sup>a</sup> The first and second dimensions indicate grid spacing in latitude and longitude, respectively. The third dimension (followed by "L") indicates the number of vertical layers extending from through the depth of the troposphere or the depth of the ocean. Numbers in square brackets denote the dimension of the global data arrays.

<sup>b</sup> Years in parenthesis indicate the period of observations used in the compilation of the statistics or the period for which the data bases are typical. Numbers not in parenthesis indicate the years for which gridded data are available.

box. The vegetation and land use data bases can be combined to represent land cover characteristics. Another vegetation data base is that of Olson and Watts [1982] and Olson et al. [1983], which is at  $0.5^\circ \times 0.5^\circ$  resolution and has  $\approx 40$  vegetation types grouped principally according to carbon density. The Wilson and Henderson-Sellers [1985] landcover data base has  $\approx 60$  cover types and resembles the Matthews data base in many respects. Bioclimatic schemes, for example, the Holdridge Life Zone Classification [1947], classify vegetation according to surface temperature, precipitation, and other climatic variables. Thus the geographic distribution of vegetation can be modeled for a given distribution of climate [e.g., Emanuel et al., 1985; Prentice, 1990].

The 1:5M FAO Soil Maps (1974-1982) are the primary source of information of soil units, soil texture, and other soil properties. The maps have been digitized independently at  $1^\circ \times 1^\circ$  resolution for the globe by Zobler [1986] and Wilson and Henderson-Sellers [1985].

Matthews and Fung [1987] compiled a  $1^\circ \times 1^\circ$  digital data base of wetland ecosystems by integrating selected components of vegetation and soils data bases with information on inundation compiled from 1:1M scale Operational Navigational Charts. While wetlands are grouped into 12 major wetland ecosystems based on characteristics important for methane emissions, the full topical detail of vegetation, soils, and inundation for the wetland sites is preserved [Matthews, 1989]. Aselmann and Crutzen [1989] independently compiled a data base on wetlands at  $2.5^\circ \times 5^\circ$  resolution.

Remote sensing provides new information about the land surface. One of the accepted parameters for global studies is the normalized difference vegetation index (NDVI), computed as the normalized difference between the near-infrared and visible radiances measured by the advanced very high resolution radiometer (AVHRR) on board polar orbiting satellites. NDVI data are available since April 1982 [Tarpley et al., 1984]. Monthly composites of the NDVI have been gridded at  $1^\circ \times 1^\circ$  resolution, compatible with the vegetation and soils data (I. Fung, unpublished atlas, 1990). The NDVI has been shown to be a good monitor of photosynthetic activity [Tucker et al., 1985, 1991; Goward et al., 1986].

*Ocean.* There is a large body of observations of sea surface temperature (SST). Global compilation of shipboard SST measurements [e.g., Alexander and Mobley, 1976] have now been succeeded by distributions which merge shipboard, buoy, and satellite measurements [Reynolds, 1982, 1988]. Historical shipboard measurements of the marine atmosphere (SST, surface air temperature, wind, pressure, humidity, and cloudiness) have been compiled into a  $2^\circ \times 2^\circ$  global grid for each month from 1854 to 1979 by the Comprehensive Ocean-Atmosphere Data Set (COADS) of Slutz et al. [1985]. Remote sensing provides global observations of SST and many other properties of the sea surface. An example is given by the atlas of Halpern et al. [1991], which presents monthly surface height, surface wind speed, and sea surface temperature for the year 1988.

Climatologies of the three-dimensional temperature, salinity, and oxygen distribution in the ocean have been compiled by Levitus [1982] from all available station data, expendable bathythermograph (XBT) data, and mechanical bathythermograph (MBT) data available at the National Oceanographic Data Center.

Phytoplankton pigment concentrations in the ocean are derived from the radiance measured by the coastal zone color scanner (CZCS) on board NIMBUS 7 [e.g., Hovis et al., 1980; Gordon et al., 1980; Esaias et al., 1986]. Data gaps exist in all the global images. The gaps result from the limited data acquisition (approximately 2 hours per day) and the problem of cloud obscuration. Because of the small space and time scales of biological processes in the ocean, there is no established algorithm for filling in the gaps.

The potential driving  $\text{CO}_2$  across the air-sea interface,  $\Delta p\text{CO}_2$ , is the difference in partial pressures of  $\text{CO}_2$  between the surface water and the air. Shipboard measurements of the partial pressure of  $\text{CO}_2$  in the surface water and in the air have been compiled and gridded at  $2^\circ \times 2.5^\circ$  resolution for different ocean basins [Takahashi et al., 1985, 1986, 1988, 1991a, b]. The data have been merged into two seasonal (January to April, July to October) distributions of  $\Delta p\text{CO}_2$  for the globe [cf. Tans et al., 1990].

### Secondary Supporting Data Bases

Secondary data bases provide geographic information on specific activities that are associated with trace gas emission, that is, they provide the  $N_{s,A_{s,t}}$  values and in some cases  $U_{A_{s,t}}$  values for equation (1). A selection of readily accessible secondary data bases is given in Table 2. Keeping these data bases current is a prerequisite to developing and maintaining accurate emission inventories. The United Nations and many nations provide annual updates on production and consumption statistics in various agricultural and energy activities. Updating the gridded data bases should be straightforward as long as political boundaries do not change and as long as the locations of the activities within the countries remain the same, although, in practice, the resources necessary to perform periodic updating are often limited or nonexistent.

Most of the secondary data bases pertain to activities in the agricultural or energy sectors, for which the principal source of data is UN statistics which give production or consumption figures for each country, or country statistics giving similar information for each state or province. Three principal data bases aid in distributing the country statistics on a  $1^\circ \times 1^\circ$  grid. The first is one of political units [cf. Lerner et al., 1988] where each  $1^\circ \times 1^\circ$  grid box is identified with a country and/or state. The second is the land use data base [Matthews, 1983] which records the dominant landuse practice in each  $1^\circ \times 1^\circ$  grid box. The third data base is that of human population density [e.g., Logan and Dignon, 1992]. The population density data set, gridded at  $1^\circ \times 1^\circ$  for the globe, was created using  $1^\circ \times 1^\circ$  global data sets on political units and landuse. After mapping individual cities with populations  $>100,000$ , the remaining population in the political unit was spread uniformly over all  $1^\circ \times 1^\circ$  grid boxes within the political boundaries that were identified with any agricultural/commercial activity other than commercial lumbering.

*Agricultural Sources.* The data bases on animal population density [Lerner et al., 1988] were based on the data bases on political units and land use. Animal population statistics for each country or state were distributed uniformly among the agricultural areas within the political unit.

TABLE 2. Existing Global Secondary Data Bases

Parameter	Spatial Resolution	Effective Year	Reference
<i>Agricultural</i>			
Agricultural cultivation	1°x1°	1980s	Matthews [1983]
Rice cultivation	1°x1°	1984	Matthews et al. [1990]
Fertilizer application	1°x1°	1984	GISS (unpublished data, 1992)
Human population	1°x1°	1980	J. A. Logan and J. Dignon, (unpublished results, 1992)
Cattle population	1°x1°	1984	Lerner et al. [1988]
Dairy cow population	1°x1°	1984	Lerner et al. [1988]
Water buffalo population	1°x1°	1984	Lerner et al. [1988]
Sheep population	1°x1°	1984	Lerner et al. [1988]
Goat population	1°x1°	1984	Lerner et al. [1988]
Camel population	1°x1°	1984	Lerner et al. [1988]
Horse population	1°x1°	1984	Lerner et al. [1988]
Pig population	1°x1°	1984	Lerner et al. [1988]
Caribou population	1°x1°	1984	Lerner et al. [1988]
<i>Industrial</i>			
Coal mines	1°x1°	1980	Seydlitz Atlas [1984]
Coal-fired power stations	Point sources	1985	Mannini et al. [1990]
Oil wells	1°x1°	1980	Seydlitz Atlas [1984]
Oil refineries	1°x1°	1980	Seydlitz Atlas [1984]
Natural gas wells	1°x1°	1980	Seydlitz Atlas [1984]

Information on the geographic and seasonal distributions of rice cultivation has been compiled and gridded at 1°x1° resolution for the globe [Matthews et al., 1991]. The geographic distribution was developed by combining landuse data identifying rice farming regimes, the data base on political units, and country statistics on annual rice harvest areas. The resultant data base gives the fraction of each 1°x1° grid box that is under rice cultivation at some time during the year. The seasonality of rice cultivation was determined using rice-cropping calendars for each rice-producing country along with statistics or estimates of the seasonal distribution of annual harvest areas in each crop cycle for the countries.

The data base on application of nitrogenous fertilizer was obtained by distributing the UN and other national statistics on fertilization consumption for each country or state among the agricultural areas within the country/state.

*Industrial Sources.* Industrial sources of trace gases are predominantly related to the production and consumption of energy. The locations and numbers of the coal mines and oil and gas wells from the Seydlitz Atlas and the Petro Atlas were directly digitized and mapped onto a 1°x1° grid. For each country/state, the production figures [U.N. Department of International Economics and Social Affairs, 1986; U.S. Department of Energy, 1986a, b] were then distributed uniformly among the production sites within the country/state.

Trace gas emission associated with the consumption of energy was assumed to be proportional to human population. Country/state statistics on natural gas consumption [U.N. Department of International Economics and Social Affairs, 1986; U.S. Department of Energy, 1986b] were distributed according to the population density within the country/state to yield a 1°x1° gridded data base on energy consumption.

### *Emission Factor Compilations*

Emission factors are the  $\Phi_{s,i,t}$  values of equation (1). The determination of reliable emission factors is difficult and exacting. It can be an engineering exercise, as in determining the flux of SO<sub>2</sub> from a specific type of ore smelting operation. It can be a plant physiology exercise, as in determining the flux of volatile organics from a specific type of vegetation. For anthropogenic processes, a notable difficulty is that some engineering modifications made to a particular source or source type are likely to require a reassessment of its emission factors.

Compilations of emission factors have been prepared for regions of the world where emissions inventory activities have been particularly detailed. We present a selection of such compilations in Table 3.

## INVENTORIES FOR GASES

### *Carbon Dioxide Inventories*

In 1973, C. D. Keeling assembled a time series of global total CO<sub>2</sub> emissions from fossil fuel burning and cement manufacture. CO<sub>2</sub> emissions for 1860 to 1953 were estimated from United Nations statistics on fuel production and U.S. Bureau of Mines data on cement production. Marland and Rotty [1984] reexamined the estimation procedure and updated the time series to 1982. In these papers the authors showed that when fuel production data can be expressed in

TABLE 3. Existing Compilations of Emission Factors

Sources Included	Compounds	Effective Year	Reference
Numerous	numerous	1985	EPA [1985]; 1987
Numerous	numerous	present	CEC [1988]
Numerous	numerous	present	CEC [1991]
Numerous	numerous	none	MHPPE [1980, 1983, 1988]
Road traffic	NO <sub>x</sub> , VOC, CO, TPM	1985	CEC [1989]
Numerous	trace elements	1982	Pacyna [1986]
Numerous	VOCs (anthro)	1985	EPA [1986]
Numerous	VOCs (anthro)	1991	EPA [1991a]
Numerous	ammonia (anthro)	1985	EPA [1990]
Numerous	biogenic HC	none	EPA [1991b]
Numerous	particulate matter (anthro)	1991	EPA [1991c]
Numerous	HCl, HF	1985	Misenheimer et al. [1985]

energy units it is possible to convert with considerable accuracy to carbon units. It is also possible to estimate the fraction of fossil fuels which are produced but not oxidized. Marland and Rotty estimated that global total anthropogenic CO<sub>2</sub> emissions values for 1958 to 1982 were accurate within 6 to 10%. This fairly low error value occurs because production was dominated by those few countries likely to have the best statistical base.

With the U.N. energy data set available on magnetic tape and covering over 200 countries or geographic regions, Marland et al. [1989] have subsequently produced a series of publications (see Table 4) estimating CO<sub>2</sub> emissions from fossil fuels and cement by country. Data are now available through 1989 and continue to be available as a continuous time series based on U.N. statistics on energy use and U.S. Bureau of Mines statistics on cement production. Individual country data are more often suspect than are the global totals. The country calculation is based on inferred consumption of fossil fuels which is taken to be production plus imports minus the sum of exports, bunker loadings, and the net of increases in stockpiles. Global average emissions coefficients are used. The sum of emissions from all countries falls short of the global total because bunker fuel loadings (fuels used in international commerce) are charged to no nation and because the individual country calculations do not try to estimate the CO<sub>2</sub> released from nonfuel uses of fossil fuels.

Using state or province-level data where available, and population density data as a proxy otherwise, Marland et al. [1985] have calculated the distribution of CO<sub>2</sub> emissions globally on a 5° by 5° grid of latitude and longitude. I. Fung

(unpublished atlas, 1990) has done similarly on a 1° by 1° grid by distributing the emission from each country/state according to the population density within the country/state. In this way, even though the population and CO<sub>2</sub> emission data are available for each 1°x1° grid cell of the globe, the effective resolution of the data sets is that of the political unit.

The other major anthropogenic source of CO<sub>2</sub> emissions is related to changes in land use that result in changes in the standing stock of biomass—most notably the clearing of tropical forests. Houghton et al. [1987] have used available data on the rate of forest clearing along with estimates of the stock and fate of carbon in those forests to derive country estimates of the rate of CO<sub>2</sub> emissions. I. Fung (unpublished atlas, 1990) distributed the Houghton et al. [1987] release figures for each country/state uniformly among the 1°x1° grid boxes of the country/state. There is a wide range of uncertainty for these values and Hall and Uhlig [1991], for example, have calculated notably smaller emissions for most countries.

An example of the interactions of models with estimates of emission fluxes is provided by the work of Tans et al. [1990]. These workers combined observed atmospheric concentrations of CO<sub>2</sub> and data on the partial pressures of CO<sub>2</sub> in surface ocean waters to identify globally significant sources and sinks of CO<sub>2</sub>. The atmospheric data were compared with boundary layer concentrations calculated with the transport fields generated by a general circulation model for specific source-sink distributions. In the model, the observed north-south atmospheric concentration gradient could be maintained only if sinks for CO<sub>2</sub> are greater in the northern than in the southern hemisphere. The observed differences between the partial pressure of CO<sub>2</sub> in the surface waters of the northern

TABLE 4. Existing Emissions Inventories: CO<sub>2</sub>

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global (fossil fuel)	country	annual	1950-1989	Marland and Boden [1991]
Global (fossil fuel)	5°x5°	none	1980	Marland et al. [1985]
Global (fossil fuel)	1°x1°	none	1980	I. Fung (unpublished atlas, 1990)
Global (biomass Comb.)	5°x5°	seasonal	circa 1982	Müller [1992]
Global (termites)	none	none	1990	Khalil et al. [1990]
Tropical (deforestation)	country	none	1980s	Houghton et al. [1987]
Tropical (deforestation)	1°x1°	none	1980s	I. Fung (unpublished atlas, 1990)
Tropical (biomass comb.)	2°x2.5°	none	1980s	Hao et al. [1990]

hemisphere and the atmosphere are too small for the oceans to be the major sink of fossil fuel CO<sub>2</sub>. Therefore Tans et al. [1990] suggest that a large amount of the CO<sub>2</sub> is absorbed on the continents by terrestrial ecosystems.

#### Carbon Monoxide Inventories

On a global scale, inventories for the major sources of CO (see Table 5) have been developed at spatial resolutions of 5° by 5° [Müller, 1992] and 1° by 1° (J. A. Logan and J. Dignon, unpublished results, 1992). For the latter, more spatially detailed inventory, the source from combustion of fossil fuels and industrial activity was determined using U.N. data for fossil fuel consumption together with emission factors for CO in a manner similar to earlier work by Logan et al. [1981] and distributing fuel use on the basis of population. CO from wood fuel was treated in similar manner to the source from fossil fuels. The source of CO from biomass burning was developed using an approach similar to Seiler and Crutzen [1980], Logan et al. [1981], and Hao et al. [1990], accounting separately for deforestation, savanna burning, agricultural waste, and wildfires. Burning was allowed only in the dry season, using a simple algorithm based on observed precipitation [Shea, 1986]. The source of CO from oxidation of isoprene allowed for the dependence of the isoprene source on temperature, sunlight, and type of vegetation (see section 4.5). The source of CO from oxidation of methane was calculated during runs of the Harvard/GISS 3-D chemical tracer model [Prather et al., 1987; Spivakovsky et al., 1990], as the source depends on local OH, and hence on CO itself.

Erickson [1989] has developed a global inventory for the ocean flux of CO with resolution of 4.5° latitude by 7.5° longitude. The source is based on a linear relationship between CO concentrations in the surface ocean and surface radiation observed during one cruise in the Atlantic Ocean [Conrad et al., 1982]. Values for surface radiation, wind and temperature fields were taken from the NCAR Community Climate Model.

More highly resolved inventories are available for CO emissions from North America, Europe and the Soviet Union, for fossil fuel and industrial sources. The NAPAP (National Acid Precipitation Assessment Program) inventory for the U.S. and Canada has the highest spatial resolution, 1/4° by 1/6° (about 18 km) [EPA, 1989]. The Soviet inventory is not

gridded, but is given in the form of emissions totals for various regions and for about 600 towns [Berlyand, 1990].

#### Methane Inventories

Total emissions inventories for methane are among the most complex of those for all the atmospheric gases, because of the large number of methane source types. Some of the sources are anthropogenic, some are natural, and many are rather poorly defined. The existing emission inventories for methane are listed in Table 6. The inventories for Eastern Europe and the USA and Canada are for industrial and automotive sources. On a global basis, fine-scale gridded data sets of all the known methane source locations have been constructed [e.g., Fung et al., 1991]. Digital data sets of methane emissions are obtained by generalizing emission factors from a few sites to broad emission classes for each source term using the primary and secondary data bases described above. Difficulties in such a generalization have been discussed by Aselman [1989]. As a result, the gridded data sets on emissions do not capture the site-to-site variation of methane fluxes observed within an emission class such as tropical swamps. They represent instead emission differences between, say, high-latitude wetlands and tropical swamps over broad geographic ranges.

Emissions estimates for all the sources must be considered at this time to be very preliminary. The best known source is perhaps emissions from domestic animals, for which annual population statistics are published. Commercial animal husbandry introduces a degree of homogenization to the population so that flux measurements may be representative of western industrialized nations. However, the largest amount of emissions are from less-developed nations where differences in feed and usage of the animals are likely to introduce large scatter in methane fluxes. Rice is likely a large source of methane. However, recent measurements show an order of magnitude differences in methane fluxes from rice within Asia. The wide scatter is not well understood, but is generally attributed to variations in practically unquantifiable factors such as fertilizers used and the history of plantings in the field. Similar uncertainties are found for methane emissions from wetlands and landfills where site-to-site variability is known to be large and the competition of the controls (such as temperature and moisture) of the emissions

TABLE 5. Existing Emissions Inventories: CO

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global (anthro, incl. biomass burning)	1°x1°	none	1980	J. A. Logan and J. Dignon (unpublished results, 1992)
Global (anthro)	5°x5°	seasonal	1986	Müller [1992]
Europe (anthro)	1°x2°	hourly	1985	Veldt [1991]
E. Europe (anthro)	150 km x 150 km	none	1982	Pacyna [1989]
N., W., and Central Europe (anthro)	1/4°x1/2°	hourly	1980, 1982	Veldt et al. [1989]
Former Soviet Union (anthro)	Republics, cities	annual	1980-1989	Berlyand [1990]
United States, Canada (anthro)	20 km x20 km	hourly (12 scenarios)	1985	EPA [1989]



TABLE 6. Existing Emissions Inventories: CH<sub>4</sub>

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global	none	none	1990	Crutzen [1991]
Global (animals)	none	none	1983	Crutzen et al. [1986]
Global (animals)	1°x1°	none	1984	Lerner et al. [1988]
Global (landfills)	continental	none	1980-1985	Bingemer and Crutzen [1987]
Global (landfills)	1°x1°	none	1984-1987	Fung et al. [1991]
Global (wetlands)	2.5°x5°	none	1990	Aselmann and Crutzen [1989]
Global (wetlands)	1°x1°	monthly	1984-1987	Fung et al. [1991]
Global (rice)	2.5°x5°	none	1980-85	Aselman and Crutzen [1989]
Global (rice)	1°x1°	monthly	1984	Matthews et al. [1991]
Global (coal prod.)	Country/ state(USA)	Yearly	1973-1987	Barns and Edmonds [1990]
Global (coal prod.)	1°x1°	none	1984	Fund et al. [1991]
Global (natural gas production)	country/ state/(U.S.A.)	Yearly	1967-1987	Barns and Edmonds [1990]
Global (natural gas consumption)	1°x1°	none	1984	Fung et al. [1991]
Global (termites)	none	none	1990	Khalil et al. [1990]
Global (termites)	none	none	1984-1987	Fung et al. [1991]
Global (biomass burning)	1°x1°	monthly	1984-1987	Fung et al. [1991]
Global (anthro)	5°x5°	seasonal	1986	Müller [1992]
Tropics (biomass burning)	2.5°x5°	none	1975-1980	Hao et al. [1990]
E. Europe (anthro)	150 km x 150 km	none	1982	Pacyna [1989]
China	country	none	1985	Khalil et al. [1991]
USA, Canada (anthro)	80 km x 80 km	hourly (12 scenarios)	1985	EPA [1989]

are not well quantified. The total methane emission from industrial sources is constrained by the fraction of <sup>14</sup>C in atmospheric CH<sub>4</sub> [Cicerone and Oremland, 1988]; the partitioning among individual source terms, however, remains uncertain. Biomass burning is another source that is highly variable in space and time, and so large uncertainties result from extrapolating from a few site measurements; its global source strength is constrained by the isotopic variation of methane in the atmosphere.

Because methane concentrations have been measured at global background sites for a number of years, and because methane's atmospheric chemistry is directly related to hydroxyl radical distributions, models and measurements can be combined to estimate the total methane emissions inventory as a function of space and time. The variation of δ<sup>13</sup>C of methane in the atmosphere provides additional constraints on the magnitudes and locations of the individual source terms. Several examples of such studies are those of Khalil and Rasmussen [1983a], Valentin and Crutzen [1990], Taylor et al. [1991], Quay et al. [1991], and Fung et al. [1991]. Model results are strongly dependent on such parameters as rate constants and OH distributions, however. In the case of methane, recent redetermination of the rate coefficient for the crucial reaction of methane with the hydroxyl radical [Vaghjiani and Ravishankara, 1991] and its <sup>13</sup>C signature [Cantrell et al., 1990] have caused a new assessment of the entire atmospheric budget of methane; the best estimate of the total emission flux to the atmosphere is now 505±105 Tg/yr [Crutzen, 1991].

#### Volatile Organic Compounds Inventories

Inventories for volatile organic compounds are very complex, not only because of the numerous source types involved, but also because the term "volatile organic compounds" (VOC) is not always given the same meaning. Species sometimes excluded are methane, ethane and (selected) CHCs and HCFCs. The term "volatile" is problematic as long as VOC species profiles are not adequately known; hence species profiles are an important part of VOC inventories. Inventories may be for anthropogenic emissions, natural emissions, or both.

VOC emissions inventories are given in Table 7. All treat anthropogenic emissions except for the studies of Lübker and Schöpp [1989], Anastasi et al. [1991], and E. Allwine et al., An initial global inventory of biogenic VOC emissions from terrestrial sources, submitted to Global Biogeochemical Cycles, 1993 (hereinafter referred to as E. Allwine et al., submitted manuscript, 1993). Two of the inventories are global [Watson et al, 1991; E. Allwine et al, submitted manuscript, 1993]. To their inherent uncertainties must be added the uncertainty resulting from the extrapolation of emission factors generated from a small number of mid-latitude regions. On a smaller scale, a similar added uncertainty from extrapolation appears in European inventories that comprise Eastern countries [Pacyna, 1989; Veldt et al., 1989; Veldt, 1991]. Inventories prepared by local experts can draw on more detailed information, at least as far

TABLE 7. Existing Emissions Inventories: Volatile Organic Compounds

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global (anthro)	10°x10°	none	1985	Watson et al. [1991]
Global (anthro)	5°x5°	seasonal	1986	Müller [1992]
Global (bio)	1°x1°	seasonal	1985	E. Allwine et al., (submitted manuscript, 1993)
Europe (anthro)	150 km x 150 km	none	1988	EMEP [1990b]
E. Europe (anthro)	150 km x 150 km	none	1982	Pacyna [1989]
EC members	admin. unit	none	1985	Bouscaren et al. [1989]
N, W, and Central	1/4°x1/2°	hourly	1980,1982	Veldt et al. [1989]
Europe (anthro, bio)				
Europe (anthro, bio)	1°x2°	hourly	1985	Veldt [1991]
OECD-Europe (anthro)	50 km x 50 km	none	1980	Lübker and de Tilly [1989]; OECD [1989,1990]
Europe (<41°E) (bio)	1°x0.5°	none	30-yr average	Lübker and Schöpp [1989]
United Kingdom (bio)	regions	none	circa 1987	Anastasi et al. [1991]
Former Soviet Union (anthro)	republics, cities	annual	1980-1989	Berlyand [1990]
United States, Canada (anthro)	80 km x 80 km	hourly (12 scenarios)	1985	EPA [1989]

as rates of anthropogenic activities are concerned [Berlyand, 1990; Bouscaren et al., 1989; EPA, 1989; Lübker and de Tilly, 1989]. The quality of these studies primarily depends on the quality of the emission factors used.

Detailed VOC profiles are incorporated into three inventories [EPA, 1989; Veldt et al., 1989; Veldt, 1991]. Broad species categories or specific substances are given by Pacyna [1989], Watson et al. [1991], and Berlyand [1990].

With respect to uncertainty, three source categories can be distinguished: (1) Technical activities that emit VOC by combustion or evaporation. (2) Land-clearing and other intentional burning. (3) Biogenic processes, i.e., generation of VOC by vegetation. To date, studies of emission factors, activity rates, and speciation profiles have concentrated on processes under category 1; however, even for these processes large uncertainties remain, mainly in the emission factors and speciation profiles. For processes in category 2, and for small-scale biomass fuel utilization, activity rates, emission factors and speciation profiles are less adequately known. Biogenic processes, in spite of much effort, are still incompletely understood.

For global research, the best that can be expected in the near future is an inventory extrapolated from studies in OECD-member countries together with additional research from other regions. In order to obtain a global inventory that addresses the same sources in all regions, compatibility of the estimation techniques to be used in the different areas must be insured.

#### *Inventories for Specific Organics or Groups*

Although "volatile organic carbon" is frequently used as a composite term for inventory purposes, it encompasses hundreds of compounds of vastly different reactivities. For that reason, inventories of specific organic compounds, or of

groups of compounds with similar characteristics, are of substantial use. Several of those that are publicly available are described in Table 8.

One of the entries in Table 8 [Watson et al., 1991] treats anthropogenic VOCs, and does so in seven groups: paraffins, olefins, common aromatics (benzene, toluene, and xylenes), formaldehyde, other aldehydes, other aromatics, and marginally reactive compounds. For these groups, U.S. emission factors were applied to the remainder of the world by use of production/consumption statistics, and the results distributed on a 10° by 10° grid based on maps of population and industrial activity. Biomass burning was also included. This approach is obviously inferior to local assessments but does provide an initial picture of grouped organic emissions for regions without local inventories, and offers several intriguing conclusions: for example, that formaldehyde emissions closely mirror industrial activity, while olefin emissions are dominated by biomass burning.

Several emissions inventories for biogenic isoprene and the terpenes are included in Table 8. The approach needed here is to determine appropriate emission rate algorithms for different types of vegetation, to apply land use classifications to provide a spatial grid of vegetative occurrence, to compile biomass density factors for vegetative types and climatic zones, and to combine all this information with temperature data to calculate emissions for each grid point. In the case of the United States and Canada, where fairly extensive field work has been done to validate the emissions algorithms, confidence in the inventories is reasonably high. For the rest of the world, and especially for tropical areas and boreal forests, much more effort is needed to improve the inventories.

Sawada and Totsuka [1986] have prepared a global emissions inventory and budget for ethylene, one of the most reactive of the hydrocarbon emittants and hence an important factor in urban atmospheric chemistry. As for the olefin group in the grouped organics inventory, the ethylene assessment

TABLE 8. Existing Emissions Inventories: Specific Organics or Groups

Compound	Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
7 Groups	global (anthro)	10°x 10°	none	1985	Watson et al. [1991]
Ethylene	global	none	none	1985	Sawada and Totsuka [1986]
Isoprene, terpenes	United States (bio)	county	seasonal	1985	Lamb et al. [1987]
Isoprene, terpenes	Canada (bio)	climatic zones/ province	seasonal	1985,88	Scholtz and Davidson, 1989
Isoprene	global (bio)	1°x 1°	none	1980	J.A. Dignon and J. Logan (unpublished results, 1990)
Isoprene, terpenes	global (bio)	1°x 1°	seasonal	1985	E. Allwine et al., (submitted manuscript, 1993)
Isoprene, terpenes	Global (bio)	5°x 5°	seasonal	1980	Müller [1992]
PAH, pesticides	Europe	150 km x 150 km	none	1982	Axenfeld et al. [1990]
PAH, pesticides	E. North America	100 km x 100 km	none	1980	Johnson et al. [1990]

shows that biomass burning is the largest global source but that anthropogenic sources are most important in urban areas.

Long-lived potentially toxic organic compounds comprise another inventory need essentially unfilled at present. The only effort to data has been an emissions survey for a number of polychlorinated hydrocarbons and polycyclic aromatic hydrocarbons in Europe [Semb and Pacyna, 1988; Duiser and Veldt, 1989; Axenfeld et al., 1990]. The emissions for major sources were calculated from data on the application of pesticides, electric power, congeneration and district heating plants, commercial, industrial, and residential combustion, transportation, and manufacturing and use of basic organic chemicals. Major parameters affecting the amount of emissions include the chemical composition of pesticides, the composition and types of fuels used, and ambient conditions, especially ambient temperature. It was concluded that sufficient data to make reliable emission estimates were available only for PAHs. The scarce and partially conflicting data for other substances allowed for only order-of-magnitude values of emissions.

#### *Oxides of Nitrogen Inventories*

The oxides of nitrogen inventories for anthropogenic sources represent one of the most thorough compilations of emissions information. The major sources of anthropogenic NO<sub>x</sub> are associated with fossil fuel burning. This class of sources primarily includes transportation, industries and utility plants. As outlined in Table 9, data bases for anthropogenic NO<sub>x</sub>, with high spatial resolution, are available for Europe and North America. Data at lower resolution is available for the former Soviet Union and Saudi Arabia. Estimates of fossil fuel burning are available for the entire globe at a lower resolution than are available for the European and North America data bases.

The accuracy of these anthropogenic NO<sub>x</sub> emissions data is somewhat less than the information for SO<sub>x</sub> emissions, but greater than for other chemicals. The greater uncertainty relative to SO<sub>x</sub> arises because of uncertainties in the NO<sub>x</sub> emissions associated with large stationary combustion sources and transportation factors. The greater certainty relative to

other chemicals arises because NO<sub>x</sub> anthropogenic sources are well known and measurements of NO<sub>x</sub> from different sources are reliable.

The greatest uncertainty with respect to the NO<sub>x</sub> emissions is the possible importance of natural sources. Preliminary estimates suggest that in some areas emissions from soils and lightning are significant [Atherton et al., 1991]. Given that detailed analysis of natural emissions is available only for North America, alternative procedures such as sensitivity studies will be needed to explore the importance of natural NO<sub>x</sub> emissions on a global scale.

#### *Nitrous Oxide Inventories*

A preliminary N<sub>2</sub>O inventory for Eastern Europe has been constructed by Pacyna [1989] (see Table 10). The first steps toward a global inventory were carried out by A. F. Bouwman et al., Global analysis of nitrous oxide emissions from natural terrestrial ecosystems, unpublished manuscript, 1992 (hereafter referred to as A. F. Bouwman et al., unpublished manuscript, 1992). In this study the potential for N<sub>2</sub>O production from natural ecosystems was determined, based on modeling. Five major regulators were used to model N<sub>2</sub>O potentials: monthly soil water and soil oxygen status (based on monthly precipitation, monthly potential evapotranspiration, soil texture, and soil drainage), carbon and nitrogen input in soils, temperature (effect of mineralization and decomposition), and soil fertility. The monthly results are organized in a 1° x 1° grid. Monthly and annual modeled N<sub>2</sub>O potentials were also compared with the available measurements to derive the grid cell emissions and regional and global totals.

Computer models can be very helpful in the case of species such as N<sub>2</sub>O whose sources are largely natural. Cicerone [1989] used atmospheric models to constrain atmospheric lifetime, global and hemispheric emissions, and interhemispheric mixing in the atmosphere. A study by Prinn et al. [1990] combines long-term atmospheric measurements of N<sub>2</sub>O with a sensitivity study to constrain the individual source strengths (fossil sources, soil and agricultural sources, biomass burning and other land transformations, and oceans).

TABLE 9. Existing Emissions Inventories: NO<sub>x</sub>

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global (anthro)	8°x10°	none	1980	Hameed and Dignon [1988]
Global (anthro)	1°x1°	none	1980	Dignon [1992]
Global (anthro)	5°x5°	seasonal	1986	Müller [1992]
Global (soil)	5°x5°	seasonal	none	Müller [1992]
Global (biomass)	1°x1°	none	1980	Dignon and Penner [1990]; Dignon et al. [1991]
Europe (anthro)	150 km x 150 km	none	1988	EMEP [1990]
Europe (anthro)	150 km x 150 km	none	1985	Pacyna et al. [1991]
EC members (anthro)	Admin. unit	none	1985	Bouscaren et al. [1989]
E. Europe (anthro)	150 km x 150 km	none	1982,1985	Pacyna [1989]
N, W, and Central Europe (anthro)	1/4° x 1/2°	hourly	1980,1982	Veldt et al. [1989]
Europe (anthro)	1°x2°	hourly	1985	Veldt [1991]
OECD-Europe (anthro)	50 km x 50 km	none	1980	Lübker and de Tilly [1989]; OECD [1989,1990]
Europe	Country	none	1985	Lübker et al. [1990]
SE Europe	Country	none	1983-1985	Katsoulis and Whelpdale [1990]
Former Soviet Union (anthro)	republics, cities	annual	1980-1989	Berlyand [1990]
Saudi Arabia (anthro)	5 regions	annual	1971-1990	Ahmed [1990]
United States, 80 km x 80 km	hourly	1985	EPA [1989]	
Canada (anthro)		(12 scenarios)		
United States, Canada (lightning, soils)	10°x10°	none	none	Fehsenfeld [1991]

TABLE 10. Existing Emissions Inventories: N<sub>2</sub>O

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
E. Europe	150 km x 150 km	none	1982	Pacyna [1989]
Global	1°x1°	monthly	1990	A.F. Bouwman et al. (unpublished manuscript, 1992)

### Ammonia Inventories

A number of regional and continental inventories have been made of agricultural NH<sub>3</sub> emissions (see Table 11). For Europe, Asman [1990] gives an update of a number of inventories for the early 1980s which also include industrial emissions (ammonia and N-fertilizer manufacture). Asman [1990] based his analysis on FAO country data (FAO Production Yearbooks) for the animal categories cattle, pigs, poultry, horses, sheep, and camels. Fertilizer data were obtained from FAO (FAO Fertilizer Yearbooks), and for each type of nitrogenous fertilizer an emission factor was used. A similar approach has been taken by Pacyna [1989] for Eastern European emissions.

The most recent study has been made by Klaassen [1990], who attempted to compute more accurate emission factors. The basis was also formed by the emission factors generally used in The Netherlands. However, for each country this factor is adapted for the nitrogen intake per animal (based on N-application and N-excretion on grasslands), and for the stall and meadow periods. The latter periods are also overall country figures. For NH<sub>3</sub> emissions from mineral fertilizers the same emission factors were used as in the work by Asman [1990].

For the United States and Canada, EPA [1989] has made a detailed inventory of all ammonia sources. It includes point sources (combustion sources, waste water treatment, refineries, ammonia synthesis, nitrogen fertilizer manufacture, and coke manufacture), mobile sources (transportation) and area sources (range animal excrement and livestock waste management and N-fertilizer applications). By far the most important sources are the agricultural area sources (unconfined and confined domestic animals). Emission factors were developed to reflect the proportion of confined versus unconfined animals (including turkeys and sheep). The emission factors used in the United States are generally lower than the ones used in the above European studies. This may be attributed to definitional differences between animal types, and also to differences in livestock management, feeding systems, etc.

### Halocarbon Inventories

Many of the halocarbons present in the atmosphere result entirely from anthropogenic sources and are manufactured worldwide by a limited number of major industrial companies. Annual production and total sales figures covering most of the

TABLE 11. Existing Emissions Inventories:  $\text{NH}_3$ 

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Europe	150 km x 150 km	none	1980	Buijsman et al. [1987]
Europe	150 km x 150 km	monthly	1988	Asman [1990]
Europe	country	none	1987	Klaassen [1990]
SE Europe	country	none	1983-85	Katsoulis and Whelpdale [1990]
E. Europe	150 km x 150 km	none	1982	Pacyna [1989]
United States,	80 km x 80 km	hourly	1985	EPA [1989]
Canada		(12 scenarios)		

world are available from various trade organizations such as the Chemical Manufacturers Association (CMA) [1986, 1990]. For  $\text{CFCl}_3$  and  $\text{CF}_2\text{Cl}_2$ , production figures by the reporting companies have an expected accuracy of better than 1% [Gamlen et al., 1986]. Atmospheric release figures, however, have an expected accuracy of approximately 5% [Gamlen et al., 1986] because of uncertainties in the storage times in various products.

Global observation networks have provided regular measurements of  $\text{CFCl}_3$ ,  $\text{CF}_2\text{Cl}_2$ ,  $\text{CH}_3\text{CCl}_3$ , and  $\text{CCl}_4$  at up to nine surface locations since 1978 [e.g., Cunnold et al., 1990; National Oceanic and Atmospheric Administration (NOAA), 1989]. By combining these observations with numerical models, annual global inventories of these gases may be estimated (see Table 12). Since the only known atmospheric destruction mechanism for  $\text{CFCl}_3$ ,  $\text{CF}_2\text{Cl}_2$ , and  $\text{CCl}_4$  is stratospheric photodissociation, and the absorption cross sections have been accurately measured, annual release rates may be estimated from the global inventories. These calculations yield satisfactory agreement with the industry's release estimates for  $\text{CFCl}_3$  and, prior to 1983, for  $\text{CF}_2\text{Cl}_2$  [Cunnold et al., 1986; Golombek and Prinn, 1986; Prather et al., 1987]. Since 1982 when the industry ceased to make estimates of production and releases in the former Soviet Union and Eastern Europe, the measurements suggest that releases in that region have remained roughly constant at about  $7 \times 10^7$  kg/yr.  $\text{CCl}_4$  is primarily used as a chemical feedstock for production of the chlorofluorocarbons; atmospheric release figures for  $\text{CCl}_4$  are however inaccurate because of the diversity of the uses of the remaining 5-10% of world production. Therefore the measured atmospheric inventory estimates are expected to yield comparable estimates of the world releases of  $\text{CCl}_4$  even though the measurements may possess a bias of approximately 25% [Simmonds et al., 1988].

$\text{CH}_3\text{CCl}_3$  releases derived from production figures have been estimated by Midgley [1989] with an accuracy of approximately 2%.  $\text{CH}_3\text{CCl}_3$  is destroyed in the atmosphere through its reaction with hydroxyl. Since the hydroxyl

distribution is poorly known, global measurements and numerical models are typically used to infer its distribution rather than to estimate  $\text{CH}_3\text{CCl}_3$  releases [Prinn et al., 1987; Spivakovsky et al., 1990; Prinn et al., 1992].

Release statistics for CFC 113, 114 and 115 have been compiled by Fisher and Midgley [1991]. Measurements of CFC 113 have been made since approximately 1985 at a few sites. At the current time the absolute calibration of these measurements is quite uncertain, but the measurements do not simulate the rapid increase in projected releases between 1985 and 1987 (P. J. Fraser, manuscript in preparation, 1991).

The spatial distribution of the releases of the halocarbons is poorly known because data on the manufacturing, shipment, and sales of the compounds are restricted by corporate confidentiality and competitive concerns. The chemical industry does break down chlorofluorocarbon sales by hemisphere and provided separate production estimates for India and Argentina until 1983. Export figures from the European Economic Community are also available. This has resulted in release estimates per semihemisphere of the globe [e.g., Cunnold et al., 1986]. However, simultaneous measurements of the halocarbons in Ireland have been interpreted [Prather, 1986] as indicating disagreement with the expected use of the halocarbons in Europe and suggesting the export of a substantial fraction of the European production. For use in numerical models, an ad hoc distribution of releases on a much finer spatial scale proportional to electric power usage has been proposed [Prather et al., 1987].

#### Sulfur Dioxide Inventories

Oxidized sulfur compounds are emitted as a result of processes where sulfur is either an unwanted impurity of various raw materials or is a raw material used to produce various goods. All these processes are quite homogeneous with respect to the sulfur emissions, with combustion of S-containing fuels being generally the predominant source.

TABLE 12. Existing Emissions Inventories: Halocarbons

Species	Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Several	global	50°x 50°	annual	1930-present	Cunnold [1990]
Several	E. Europe	150 km x 150 km	none	1982	Pacyna [1989]
$\text{CH}_3\text{CCl}_3$	global	latitude bands	annual	1970-1988	Midgley [1989]
$\text{CCl}_4$	United States	none	annual	1930-present	Cunnold [1990]
CFC 113, 114, 115	global	none	annual	1980-1988	D.A. Fisher and P. Midgley (unpublished manuscript, 1991)

Many reasonably good SO<sub>2</sub> inventories, in fact, are constructed merely by determining the amount of coal used, and its sulfur content.

For no species emitted to the atmosphere has more effort gone into emissions studies than is the case for SO<sub>2</sub>. As a result, emission estimates based on sulfur content of fuels, both fossil and biomass, and wastes are relatively accurate. A major effort to estimate SO<sub>2</sub> emissions in the United States and Canada was made within the National Acid Precipitation Assessment Program (NAPAP) with results published by the EPA [1989]. In Europe, emission data have been compiled within various programmes, as indicated in Table 13. In some cases, the spatial resolution is quite high. Particularly detailed inventories of SO<sub>2</sub> emissions exist as well for Saudi Arabia, the USSR, and Southeast Asia.

Emission factors for SO<sub>2</sub> are well established. If sufficient effort has gone into determining the amount of fossil fuel consumed, the sulfur content of that fuel, and the degree of activity of other important sources such as ore smelting, the resulting emissions inventories can be of very high quality. However, two major uncertainties often exist. One is that for a given fuel type the average sulfur content varies from country to country by more than a factor of 3. Data on the sulfur content of fuels and ores in many parts of the world are lacking. The second uncertainty has to do with the degree of recovery of sulfur from industrial and combustion processes. In many regions of the world little sulfur is recovered during combustion, yet varying degrees are captured during ore smelting and petroleum refining.

Finally, two natural SO<sub>2</sub> sources should be mentioned. One is volcanoes, which appear to account for about 10% of the global source flux of atmospheric sulfur on average [Stoiber et al., 1987]. The second is biomass burning, which is surely of only secondary importance on a global basis but

could control the sulfur budget over large regions of the tropical continents during the burning seasons [Andreae et al., 1988].

### *Reduced Sulfur Inventories*

The increased awareness of the environmental consequences of acidic precipitation has resulted in many recent measurements of the reduced sulfur gases emitted to the atmosphere from the ocean and terrestrial biosphere. These additional data have been used to re-assess earlier global natural sulfur emission estimates [Andreae, 1986; Khalil and Rassmussen, 1984] and to further refine these estimates on regional spatial scales and seasonal time scales [Guenther et al., 1989; Erickson et al., 1990; Bates et al., 1992; Spiro et al., 1992]. The existing emission inventories are summarized in Table 14.

Guenther et al. [1989] used emission algorithms based on the data collected by Lamb et al. [1987] to develop a U.S. national emissions inventory with a county spatial scale and monthly time scale. Information from the Geoecology Data Base [Olson, 1980] and U.S. Geological Survey National Atlas [1970], was used to determine surface areas of wetland, organic and other soils, croplands, natural vegetation, and inland waters for all counties in the contiguous United States. Deciduous and coniferous forest surface area estimates were converted to leaf biomass estimates using the relationships developed by Zimmerman [1979].

These same algorithms were used by Bates et al. [1992] in a global sulfur emission inventory. The inventory was divided spatially into 15° latitude belts and temporally into two seasons. Terrestrial biogenic sulfur emissions (COS, CS<sub>2</sub>, DMS, DMDS, H<sub>2</sub>S) were calculated using climatic data compiled on a 2° by 2° spatial scale and monthly temperature

TABLE 13. Existing Emissions Inventories: SO<sub>2</sub>

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global (anthro)	8°x10°	none	1980	Hameed and Dignon [1988]
Global (anthro)	1°x1°	none	1980	Dignon [1992]
Global (anthro)	5°x5°	seasonal	1986	Müller [1992]
Global	1°x1°	monthly	1980	Spiro et al. [1992]
Europe (anthro)	150 km x 150 km	none	1988	EMEP [1990a]
EC members (anthro)	admin. units	none	1985	Bouscaren et al. [1989]
E. Europe (anthro)	150 km x 150 km	none	1982,85	Pacyna [1989]
N, W, and Central Europe (anthro)	1/4°x 1/2°	hourly	1980,1982	Veldt et al. [1989]
Europe (anthro)	1°x2°	hourly	1985	Veldt [1991]
OECD-Europe (anthro)	50 km x 50 km	none	1980	Lübker and de Tilly [1989]; OECD [1989,1990]
Europe	Country	none	1985	Lübker et al. [1990]
SE Europe	Country	none	1983-1985	Katsoulis and Whelpdale [1990]
Former Soviet Union (anthro)	republics, cities	annual	1980-1989	Berlyand [1990]
East Asia	80 km x 80 km	none	circa 1986	Fujita et al. [1991]
Saudi Arabia (anthro)	5 regions	annual	1971-1990	Ahmed [1990]
China	province	none	1985	Wenxing and Quan [1990]
United States, Canada (anthro)	80 km x 80 km	hourly	1985	EPA [1989]
		(12 scenarios)		

TABLE 14. Existing Emissions Inventories: Reduced Sulfur

Compound	Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
All	global	none	none	1985	Andreae [1986]
All	global	latitude bands	seasonal	1980	Bates et al. [1992]
H <sub>2</sub> S, DMS	global	1°x1°	monthly	1980	Spiro et al. [1992]
DMS	global	4.5°x7.5°	seasonal	none	Erickson et al. [1990]
All	United States	county	monthly	1985	Guenther et al. [1989]
COS, CS <sub>2</sub>	hemispheric	none	none	1980	Khalil and Rasmussen [1984]

scale by Shea [1986] and land cover data compiled by Wilson and Henderson-Sellers [1985] on a 1° by 1° grid. The primary (> 50% of the grid) and secondary (25% to 50% of the grid) land cover types were compiled into 12 simple surface types by latitude band. Seasonal emission rates were calculated for five sources: cropland vegetation, wetland, deciduous and coniferous canopies, and soils (total soil area = natural vegetation area + agricultural land areas + bare soil area). Marine sulfur emission were calculated using measured seawater dimethylsulfide concentrations and air-sea exchange models [Bates et al., 1987; Liss, 1973]. The emissions were extrapolated regionally and seasonally from the North Pacific Ocean to the other oceans.

The total flux of biogenic reduced sulfur to the atmosphere is calculated at 0.5 Tmole/yr [Bates et al., 1992] and is almost entirely (>95%) from the oceanic emission of DMS. The uncertainty in the ocean flux of DMS is approximately a factor of two.

#### Hydrogen Chloride Inventories

Emissions inventories for HCl are at present in a rather confused and incomplete state. Many anthropogenic sources are known, coal combustion and waste incineration being by far the most important. The emission factors are quite variable, and a large uncertainty factor should be assigned to any anthropogenic flux estimates. For marine or coastal regions, however, any such uncertainties are dwarfed by estimates that HCl volatilization from sea salt may be as much

as 100–400 Tg/yr [Singh and Kasting, 1988; Möller, 1990], perhaps a hundred times the anthropogenic source on a global basis.

The available regional inventories for anthropogenic HCl are listed in Table 15. In each case, the spatial resolution is by political boundaries. It is probably appropriate to regard the values provided as order-of-magnitude estimates only.

#### Hydrogen Fluoride Inventories

Only one emissions inventory is known for HF; it treats emissions from large industrial sources in the United States and Canada (see Table 16). Misenheimer et al. [1985] compiled and rated emission factors available from the literature for coal combustion, hydrogen fluoride manufacture, the primary aluminum industry, and the phosphate fertilizer industry. These were then applied to plant throughput values reported to the EPA [1989]. (Since not all facilities reported throughput, the inventory is not complete.) No information is presented for small, dispersed sources of HF. The inventory reports that the most significant point source of HF in the United States is coal-fired utilities.

#### Radionuclide Inventories

Radon 222 is a daughter of <sup>226</sup>Ra, which is an ubiquitous constituent of crustal materials. Emission of <sup>222</sup>Rn to the atmosphere follows from radioactive decay of <sup>226</sup>Ra in soil

TABLE 15. Existing Emissions Inventories: HCl

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
United States, Canada (anthro)	state, province	hourly (12 scenarios)	1985	EPA [1989]
W. Europe (anthro)	country	none	1985	Lightowlers and Cape [1988]
Europe (anthro)	country	none	1980	Möller [1990]

TABLE 16. Existing Emissions Inventories: HF

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
United States	State	hourly (12 scenarios)	1985	EPA [1989]
Canada	province	hourly (12 scenarios)	1985	EPA [1989]

and upward transport of  $^{222}\text{Rn}$  through the soil gas to the surface. The flux of  $^{222}\text{Rn}$  depends on the  $^{226}\text{Ra}$  abundance in soil, on the physical properties of the soil, and on meteorological conditions. Estimates of the global mean  $^{222}\text{Rn}$  flux from soils range from  $0.75 \text{ atoms cm}^{-2} \text{ s}^{-1}$  to  $1.2 \text{ atoms cm}^{-2} \text{ s}^{-1}$ . Emissions from water surfaces are two orders of magnitude lower and can be neglected as a source of  $^{222}\text{Rn}$  over continents.

The mean emission flux is altered by changes in surface pressure and by the presence or absence of freezing conditions. Adjustments to the mean flux to take these factors into account form the basis for the preliminary inventories (Table 17) of Jacob and Prather [1990] and Hameed and Dignon [1990]. Other factors that should be considered include the  $^{226}\text{Ra}$  content of the soils, the soil moisture, and vegetative cover. Efforts are in progress by Dörr and Munnich [1990, also manuscript in preparation, 1992], but much experimental and theoretical work remains to be accomplished before reliable radon inventories are available.

Krypton 85 (half-life 10.76 years) is emitted during reprocessing of nuclear fuel. There are only a few important sources of  $^{85}\text{Kr}$ , all in the northern hemisphere and all confined to a narrow band of latitudes between  $33^\circ$  and  $52^\circ\text{N}$ . Emissions from western sources, mainly in the United Kingdom, France, and the United States, are well documented [Von Hippel et al., 1986]. The source from the Soviet Union has been inferred by simulating observed atmospheric concentrations with a general circulation model [Jacob et al., 1987].

Beryllium 7 (half-life 53 days) is produced in the stratosphere and in the upper troposphere by cosmic ray spallation reactions with nitrogen and oxygen. Estimates of the source as a function of latitude and altitude have been presented by Bhandari et al. [1966] and O'Brien [1979]; the global estimates differ by 33% but the geographical

distributions are similar. The production of  $^7\text{Be}$  is a maximum in polar regions at 25-30 km altitude. 30% of the total source is in the troposphere.

Unlike most of the other inventories discussed in this paper, those for the radionuclides are highly model-related. That is, unlike the rigorous approach described by equation (1), in which specific values are assigned to parameters related to source abundances and strengths, an atmospheric circulation model is combined in some way with observations or with relatively unconstrained assumptions to infer source strengths. Such approaches provide a good starting point for inventory development, but should be recognized as first approximations to be improved as additional location-dependent source information is acquired.

## INVENTORIES FOR AIRBORNE PARTICULATE MATTER

### Total Particulate Matter

Emissions inventories for total particulate matter are given in Table 18. The airborne particulate emissions inventory developed by the EPA [1989] includes emissions from anthropogenic sources (e.g., industrial facilities, power plants, motor vehicles, and furnaces), as well as those from wind erosion, dust devils (intense convective wind events), and unpaved roads. Estimates of the alkalinity content and the size distribution of the particulate matter are included; these considerations affect the reactivity of the particulates, their potential transport distance, and their lifetime in the atmosphere. Smaller particles usually travel longer distances and have longer atmospheric lifetimes.

Since the EPA inventory was developed for purpose of analyzing the acidic deposition phenomenon, a method was

TABLE 17. Existing Emissions Inventories: Radionuclides

Species	Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
$^{222}\text{Rn}$	global	$1^\circ \times 1^\circ$	none	NA	Hameed and Dignon [1990]
$^{222}\text{Rn}$	global	$8^\circ \times 10^\circ$	none	NA	Dörr and Munnich (unpublished data, 1992)
$^{222}\text{Rn}$	N. Amer.	$4^\circ \times 5^\circ$	none	NA	Jacob and Prather [1990]
$^{222}\text{Rn}$	Germany	none	none	NA	Dörr and Munnich [1990]
$^{85}\text{Kr}$	global	Point sources	annual	1945-1983	Von Hippel et al. [1986]; Jacob et al. [1987]
$^7\text{Be}$	global	Lat, alt	NA	NA	Bhandari et al., 1966
$^7\text{Be}$	global	Lat, alt	NA	NA	O'Brien, 1979

TABLE 18. Existing Emissions Inventories: Total Particulate Matter

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global	latitude bands	none	1990	Penner et al. [1990]
Former Soviet Union (anthro)	republics, cities	annual	1980-89	Berlyand [1990]
United States, Canada (TPM)	state, province	hourly (12 scenarios)	1985	EPA [1989]
United States (alkaline)	U.S. counties	none	1985	Barnard [1990]



developed [Barnard, 1990; Gillette and Sinclair, 1990] to estimate alkaline particle emissions, since those particles can neutralize acids in the atmosphere. Due to the difficulty in determining the amount of dust that remains airborne, the elemental content of the particulates, and the sporadic nature of wind action sources, these estimates of wind-borne alkaline dust are model-dependent and subject to high levels of uncertainty.

Estimated emissions of particles in the former Soviet Union from industrial sources are available from Berlyand [1990]. Industrial emissions of particles for individual cities and towns, as well as for broad economic regions and the former Soviet Union as whole were estimated. The inventory includes estimates of emissions of specific particles including lead, organic and inorganic dust, various metal compounds, fluorine dust, cement dust, and others. The change in emissions between 1985 and 1989 is also included. No size distributions are available.

#### *Sulfate Particulate Inventories*

For the estimation of the anthropogenic contribution to acidic or total sulfate deposition it is important to have data on fractions of SO<sub>2</sub> emissions that are converted to sulfate particulates before they enter the atmosphere. NAPAP provides emission factors for these components for a number of sources [EPA, 1989]. In general, the PHOXA project [Veldt et al., 1989; Veldt, 1991] used percentages of SO<sub>2</sub> emission factors, based on a literature review [e.g., Goklany et al., 1984]. At present, these inventories (see Table 19) must be regarded as only very rough estimates.

#### *Trace Metals Inventories*

The first quantitative assessments of worldwide emissions of trace metals from natural and anthropogenic sources have

been prepared during the 1980s (see Table 20). In the study by Nriagu and Pacyna [1988], emission factors and the statistics on global production and consumption of industrial goods have been used to calculate the worldwide emissions of 16 trace metals from major anthropogenic sources. Annual anthropogenic fluxes were calculated to be circa 18.8 Gg As, 7.6 Gg Cd, 33.4 Gg Cu, 3.6 Gg Hg, 55.7 Gg Ni, 330 Gg Pb, and 130 Gg Zn. A comparison of these fluxes with the natural emissions leaves no doubt as to the influence of industrial activities on the atmospheric cycle of the trace metals. On average the anthropogenic emissions of As, Cd, Cu, Ni, and Zn exceed the inputs of these metals from natural sources by about two-fold or more; in the case of lead the ratio of anthropogenic to natural emission rates is about 17.

Combustion of hard coal, lignites and brown coal in electric power plants and in industrial, commercial and residential burners is the major source of airborne Hg, Mo, and Se and a very significant source for As, Cr, Mn, Sb, and Tl. Combustion of oil for the same purpose is the most important source of V and Ni and is an important contributor of Sn. The non-ferrous metal industry accounts for the largest fraction of Pb (in addition to gasoline combustion), As, Cd, Cu, and Zn emitted.

The spatial distribution of trace element emissions is not available on a global scale. Within Europe and North America, however, reasonable but varied spatial resolution has been achieved, and a speciated mercury inventory is available for North America.

#### *Soot Inventories*

Soot particles are injected into the atmosphere as a result of the combustion of fossil fuels and by the large-scale burning of biomass. Soot particles in the atmosphere may be important to the global climate directly by the scattering and absorption of sunlight [Charlock and Sellers, 1980] and indirectly by acting as cloud condensation nuclei. Twomey

TABLE 19. Existing Emissions Inventories: Sulfate Particulates

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
N, W, and Central Europe (anthro)	1/4° x 1/2°	hourly	1980,1982	Veldt et al. [1989]
Europe (anthro)	1° x 2°	hourly	1985	Veldt [1991]
United States, Canada (anthro)	state, province	hourly (12 scenarios)	1985	EPA [1989]
North America	127 km x 127 km	none	1985	Voldner and Smith [1990]

TABLE 20. Existing Emissions Inventories: Trace Metals

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global	no resolution	none	1983	Nriagu and Pacyna [1988]
Europe	150 km x 150 km	none	1982	Pacyna and Munch [1988]
Europe	industrial regions	none	1980	Pacyna [1984]
United Kingdom	country	none	1983	Hutton and Symon [1986]
United States, Canada	state, province	hourly (12 scenarios)	1985	EPA [1989]
North America	127 km x 127 km	none	1985	Voldner and Smith [1990]

[1977] has discussed the effect of cloud condensation nuclei altering the microphysical and radiative processes of clouds and their albedo.

No direct measurements exist for the emissions factors of soot particles from biomass burning. However, Radke et al. [1988] have reported emissions factors and optical absorption coefficients for all particles, and Hao et al. [1989] has provided an inventory for the amount of biomass burned. By combining this information, Penner et al. [1990] have produced a model-related global emissions inventory of soot particles (see Table 21). The inventory is divided into latitude bands, is highest in the tropical areas where biomass burning predominates, and estimates a global biomass burning soot flux of about 7 Tg C/yr.

The anthropogenic source of soot aerosol from fossil fuel combustion has been estimated by Penner et al. [1992]. This inventory, clearly a first attempt that will benefit from more extensive field measurements, suggests a global anthropogenic soot flux of about 25 Tg C/yr, several times that due to biomass burning. However, most of the anthropogenic source is in the northern hemisphere and much of the biomass burning is in the tropics or southern mid-latitudes, so the source of soot particles differs with latitude band.

## DISCUSSION

It is apparent from the wide variations in types of emissions inventories and their properties that inventories can be constructed in many ways and with many purposes in mind.

One way in which a standard level of uniformity is reached is for a single inventory to deal simultaneously with several major species. Table 22 lists those that have been mentioned here.

It is appropriate not only for us to cite existing emissions inventories and their characteristics, but also for us to assess where additional efforts are needed. For this purpose, we have assigned in Table 23 qualitative ratings to emissions inventories for all species or groups of species we have discussed. The basis for the ratings is the degree of availability of emissions inventories and estimates by experts of the level of uncertainty in these inventories. The ratings are obviously subjective, but demonstrate that the only species for which global fluxes are regarded as well known are the CFCs. The overall spatial resolution of the inventories is uniformly poor or nonexistent. For specific regions, however, the spatial resolution of the emissions information can be regarded as good for CO<sub>2</sub>, NO<sub>x</sub>, and SO<sub>2</sub>. The temporal resolution of the inventories is almost universally fair, poor, or nonexistent. The exception is CO<sub>2</sub> in some regions. This does not imply superior time resolution for CO<sub>2</sub> inventories, but rather that for most purposes annual information for CO<sub>2</sub> emissions is sufficient. Except for most of North America and Western Europe, detailed regional inventories are available for very few species and very few regions. All in all, we find that present emissions inventories are unsatisfactory in one way or another for every species surveyed, a caution that users of inventory data must keep always in the forefront of their thinking.

TABLE 21. Existing Emissions Inventories: Soot

Region	Spatial Resolution	Temporal Resolution	Effective Year	Reference
Global	latitude bands	none	1990	Penner et al. [1990]
Global	none	none	1990	Penner et al. [1992]

TABLE 22. Emission Inventories Dealing With Several Species

Species	NAPAP <sup>(a)</sup>	NILU <sup>(b)</sup>	PHOXA <sup>(c)</sup>	FSU <sup>(d)</sup>	EMEP <sup>(e)</sup>	OECD <sup>(f)</sup>	CITEPA <sup>(g)</sup>
SO <sub>x</sub>	x	x	x	x	x	x	x
NO <sub>x</sub>	x	x	x	x	x	x	x
VOC	x	x	x	x	x	x	x
CH <sub>4</sub>	x	x					
CO	x	x	x				
NH <sub>3</sub>	x	x			x		
HCl	x						
HF	x						
CFCs							
Particles	x			x			
SO <sub>4</sub> <sup>2-</sup> particles	x		x				
Trace metals	x	x					

<sup>(a)</sup> Information contained in EPA [1989].

<sup>(b)</sup> Information contained in Pacyna [1989].

<sup>(c)</sup> Information contained in Veldt et al. [1989]; Veldt [1991].

<sup>(d)</sup> Former Soviet Union; information contained in Berlyand [1990].

<sup>(e)</sup> Information contained in EMEP [1990]a,b.

<sup>(f)</sup> Information contained in OECD [1989] [1990].

<sup>(g)</sup> Information contained in Bouscaren et al. [1989].

TABLE 23. Status Assessments of Emission Inventories

Species	Global Flux	Spatial Resolution		Temporal Resolution	
		Specific Regions	Overall	Specific Regions	Overall
CO <sub>2</sub>	F	G	P	G	G
CO	F	G	P	F	NI
CH <sub>4</sub>	F	F	P	P	P
VOC	P	F	P	F	P
PAH	P	P	NI	NI	NI
Chlorinated HC	P	P	NI	NI	NI
NO <sub>x</sub>	F	G	P	F	P
N <sub>2</sub> O	P	F	P	F	F
NH <sub>3</sub>	NI	F	NI	F	NI
CFC	G	P	P	P	F
SO <sub>2</sub>	F	G	P	F	P
Reduced S	F	F	P	F	P
HCl	NI	F	NI	NI	NI
HF	NI	P	NI	NI	NI
Radon	F	P	P	NA	NA
TPM	P	F	P	NI	NI
SO <sub>4</sub> <sup>2-</sup>	NI	P	NI	P	NI
Metals	P	P	NI	NI	NI
Soot	P	P	P	NI	NI

G=good; F=fair; P=poor; NI=no inventory; NA=not applicable.

The research reviewed in this paper has dealt primarily or exclusively with inventories for the present time and under normal conditions. For completeness, we note here that three other types of inventories are often of use. They are (1) historical emission inventories, in which inventories for one or more epochs in the past are estimated; (2) special occurrence emission studies, in which emissions from such events as volcanic eruptions, bolide impacts, and forest fires are assessed or estimated; and (3) future emission scenarios, in which projections of such factors as populations, industrial activity, and emission control technology are used to predict emissions in future epochs. Reviewing inventories such as these and creating new special occurrence inventories as needed are among the anticipated activities of the GEIA group.

The availability of the inventories presented in this paper varies widely. The references cited are themselves insufficient, since generally the full list of an inventory's numerical values is not suitable for publication and can be acquired only by direct contact with the author. The GEIA group does not maintain or distribute these inventories. However, the reference list given here should aid in interactions and data transfer among researchers. In addition, as the GEIA group develops and validates its own inventories through the efforts of international, intercontinental project teams, those inventories will be available to the entire scientific community in a variety of electronic and print forms, and summaries will be published in the scientific literature.

To summarize, truly useful and accurate emissions inventories are those produced by environmental accountants, process engineers, natural scientists and computer modelers working in tandem. Alternatively, efforts involving a subset of these specialties benefit from knowledge of and close ties to the others. It is this unity of effort that is central to the GEIA/IGAC emissions inventory activity.

**Acknowledgments.** Inventory development is a demanding task, one that is never finished, and one that frequently does not get the credit it deserves. We applaud those who have constructed the data bases and inventories cited herein, and encourage them to continue to produce inventories of the highest possible accuracy and utility. For communicating inventory information to us, we thank M. E. Berlyand, R. Bouscaren, W.-M. Hao, B. Lübkert, C. M. Spivakovsky, K. M. Valentin, E. C. Voldner, and W. Wenxing.

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